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Density scaling and quasiuniversality of flow-event statistics for athermal plastic flows

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Athermal steady-state plastic flows were simulated for the Kob-Andersen binary Lennard-Jones system and its repulsive version in which the sign of the attractive terms is changed to a plus. Properties evaluated include the distributions of energy drops, stress drops, and strain intervals between the flow events. We show that simulations at a single density in conjunction with an equilibrium-liquid simulation at the same density allow one to predict the plastic flow-event statistics at other densities. This is done by applying the recently established “hidden scale invariance” of simple liquids to the glass phase. The resulting scaling of flow-event properties reveals quasiuniversality, i.e., that the probability distributions of energy drops, stress drops, and strain intervals in properly reduced units are virtually independent of the microscopic pair potentials.

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Solids do not flow according to the traditional definition [1,2]. Actually a solid *does* flow at any finite stress, in principle, even at low temperatures [3,4]. Finite-stress flow of a solid is referred to as plastic flow [5–11]. Such flows are observed in practice, e.g., when a piece of metal is deformed, but solids are generally useful in real life, of course, precisely because they do *not* plastically deform under normal circumstances.

In science, plastic flow is often monitored by subjecting the solid in question to a shear deformation that increases linearly in time—if the solid does not break, it eventually yields and flows [12]. In the longer run a steady state is reached in which the shear stress fluctuates around an average [13]. This paper focuses on the steady-state plastic-flow properties of amorphous materials, which are relevant, e.g., for applications of metallic glasses [14,15]. In this field the shear-transformation-zone [16–19] and the soft-glassy-rheology [20–22] theories have been successful in explaining the rich phenomenology observed in elastoplastic flows and the yielding transition. Supplementing these approaches, mode-coupling theory has been applied with a focus on constructing a first-principles theory of yielding [23–25]. We here supplement these successful theories by showing that the plastic-flow statistical properties at one density uniquely determine those at other densities. This is done by extending to glasses a recently established scaling property [26,27] of the so-called “Roskilde-simple” liquids [27–36].

Plastic flows can take place even at zero temperature [3,4]. For an amorphous solid like a metallic glass the zero-temperature plastic flow properties provide important information about the energy landscape of the atoms [3,4,37]. For this reason athermal flows of amorphous solids have recently been studied extensively by computer simulations [13,38–41]. Much of the focus has been on the interesting finite-size scaling properties of the flow-event statistics [38,42–45], and by now a good understanding has been achieved of the general nature of the self-organized criticality observed in steady-state plastic flows [46].

Plastic flows take place as the result of large stresses. In practice, these are rarely pure shear stresses, but involve also locally high pressures or, occasionally, even negative pressures. It is therefore important to understand the effect of pressure on a plastic flow, and this subject has indeed been investigated intensively over the years. Thus, it is well known that the rate of plastic flow decreases dramatically with increasing pressure [47,48], typically following an exponential function, a fact that is often rationalized in terms of the free-volume model [49].

In the past decade experiments on *supercooled liquids* have shown that if a simple description is aimed at, the right quantity to focus on is not the pressure but the density. Thus, for a large number of glass-forming liquids in metastable equilibrium it has been demonstrated that the viscosity—or, equivalently, the average relaxation time—is a function of ρ^γ/T in which $\rho = N/V$ is the number density, T the temperature, and γ the so-called density-scaling exponent (not to be confused with the shear displacement that is traditionally also denoted by γ). This so-called power-law density-scaling relation was established by Alba-Simionesco and coworkers, as well as by Roland, Paluch, and coworkers, for many van der Waals bonded organic liquids and polymers [50–53]. For large density changes the density-scaling exponent is not constant and a more general form of scaling takes over [50,54,55], which is the one used below.

One purpose of the present paper is to show that the insight gained from the study of supercooled liquid dynamics—that density rather than pressure is the important variable—can be used for understanding the scaling properties of athermal plastic flows. We develop a theory of the density dependence of the statistics of athermal plastic flow events at steady state, which from the properties at one density makes it possible to predict the steady-state behavior at any other density (thus, the history dependence of the initial flow is not under study). A first step in this direction was taken in Ref. [56] by assuming a constant, empirically determined exponent for the density scaling of the average properties of a stationary plastic flow, a procedure that is analogous to the above-mentioned power-law density scaling for liquids. A more general scaling is established below based on the concept of “hidden scale invariance” [27]. The second purpose of this paper is to show

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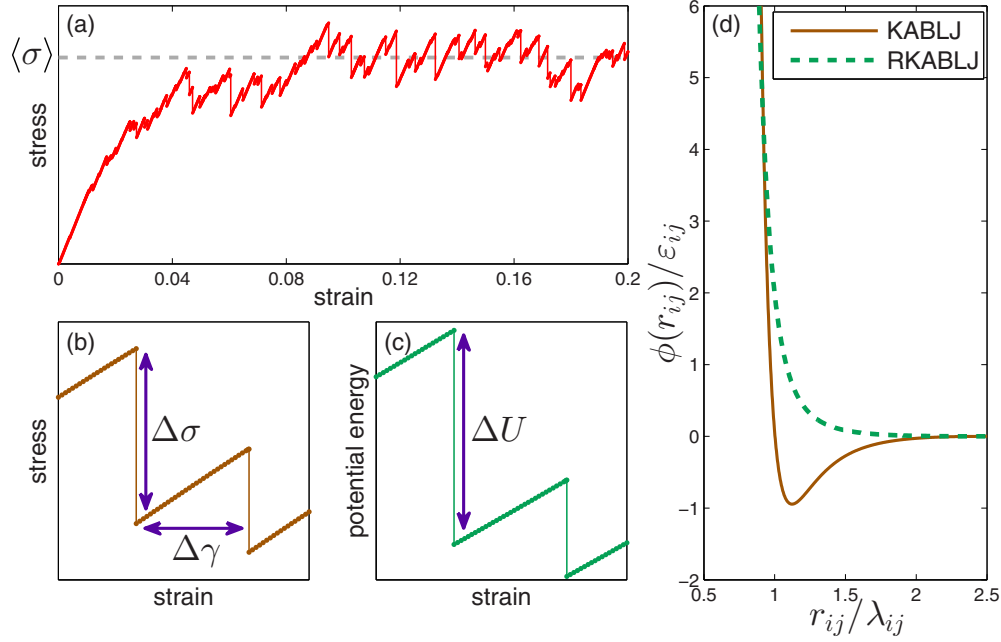


FIG. 1. (Color online) (a) Typical stress-strain signal for a glass that is shear deformed under athermal quasistatic conditions. The horizontal dashed line indicates the steady-state average flow stress $\langle \sigma \rangle$. (b), (c) Illustration of the key observables measured in this work: the (shear) stress drops $\Delta \sigma$, the strain intervals between drops $\Delta \gamma$, and the energy drops ΔU . (d) The Lennard-Jones pair potential of the Kob-Andersen binary Lennard-Jones (KABLJ) model and its repulsive counterpart with a plus in front of the r^{-6} term (RKABLJ) as functions of the distance between particles i and j , r_{ij} .

that by using this novel scaling, which involves no more parameters than the traditional power-law scaling, “quasi-universality” [57] of the flow-event properties is revealed. By this we mean that in properly scaled units the flow-event statistical properties are (almost) independent of the microscopic interactions.

Simulations of zero-temperature flows of simple model systems were performed in order to investigate the effect of density. Zero temperature (“athermal”) means that the system is always at a local minimum in the energy landscape—thus no atom exhibits any thermal vibration. In the simulations each system was subjected to a quasistatic shear deformation, corresponding to a very low strain rate. Every now and then the system exhibits a sudden plastic flow event [38], an avalanche that takes it from a mechanically marginally stable state, i.e., one with a zero eigenvalue of the potential-energy Hessian, to a mechanically stable state.

A typical stress-strain signal following shear deformation is displayed in Fig. 1(a). Starting from the as-quenched glass the stress initially increases linearly and few flow events take place. This is the standard linear, elastic response of a solid to an imposed shear deformation. At long times a fluctuating steady state is reached in which the stress saturates. Here, over short time spans the stress increases linearly and continuously with deformation, but drops discontinuously whenever a flow event takes place. As mentioned, this happens when a mechanically marginally stable state is reached, at which point the system tumbles down the potential-energy landscape to a new state of mechanical equilibrium. From here, the system is gradually shear deformed, behaving like a continuously strained elastic solid, and the shear stress increases until a

new flow event takes place at which point the stress decreases abruptly, etc.

Following previous works in the field [38,40,42,44,58] we probed the statistics of the plastic flow events by determining the probability distributions of the flow events’ shear-stress drops and energy changes, as well as the strain intervals between consecutive flow events [Figs. 1(b) and 1(c)]. Specifically, we simulated two model glass-forming systems in three dimensions, each consisting of particles of two sizes. Single-component systems were not studied because, even if prepared into an amorphous state by rapid quenching from the liquid, they crystallize when subjected to plastic deformations and no steady state of a glass plastic flow can be reached [59]. The interaction potentials simulated are the standard Kob-Andersen binary Lennard-Jones (KABLJ) system consisting of 80% large and 20% small particles [60] and its repulsive counterpart (RKABLJ) [61]. The LJ pair potential between particles i and j is the usual $4\epsilon_{ij}[(r_{ij}/\lambda_{ij})^{-12} - (r_{ij}/\lambda_{ij})^{-6}]$, while the RLJ pair potential is $(\epsilon_{ij}/2)[(r_{ij}/\lambda_{ij})^{-12} + (r_{ij}/\lambda_{ij})^{-6}]$ (the KABLJ pair potential follows the standard LJ normalization of having minimum energy $-\epsilon_{ij}$, whereas the RKABLJ potential is normalized by requiring the energy to be ϵ_{ij} at $r_{ij} = \lambda_{ij}$ [61]). The particle masses do not enter into the problem. The AQS scheme for athermal plastic-flow simulations was used [38,42,44,62], which consists of repeated applications of incremental shear-strain deformations followed by a minimization of the potential energy using a standard nonlinear conjugate-gradient algorithm.

Each system simulated consisted of $N = 8000$ particles. We first deformed the system under athermal shear as described above to a strain of ~ 1 . This was done in order to assure that

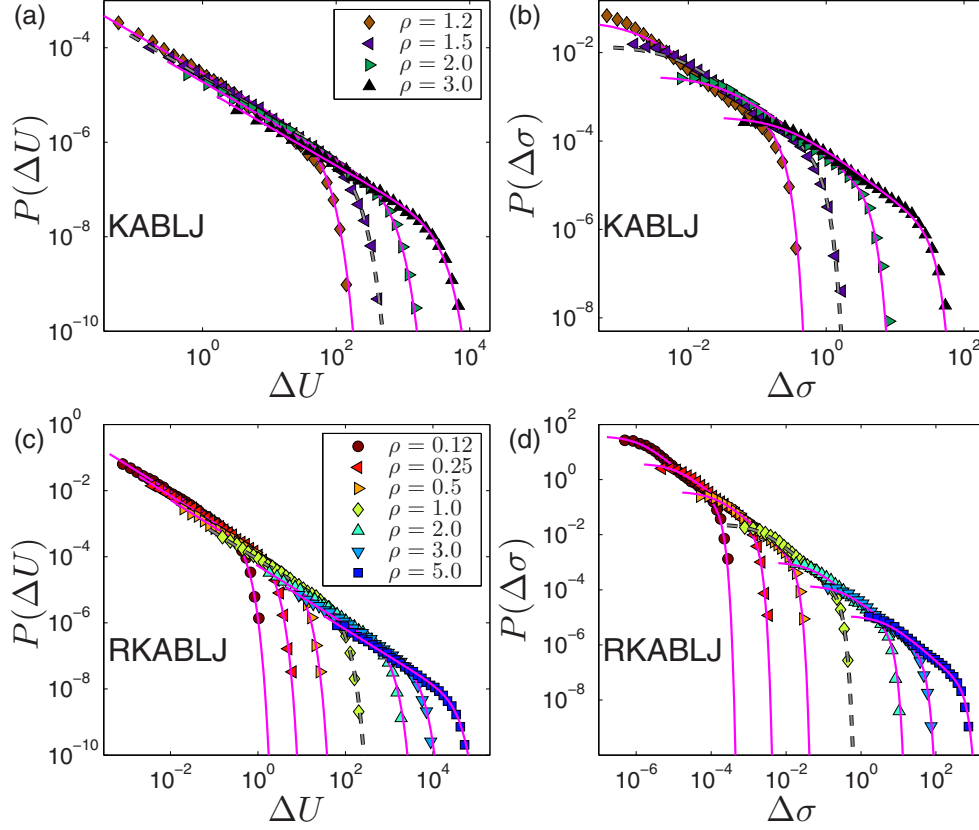


FIG. 2. (Color online) Distributions of potential-energy drops $P(\Delta U)$ for (a) the KABLJ and (c) the RKABLJ systems at different densities, and distributions of stress drops $P(\Delta \sigma)$ for (b) the KABLJ and (d) the RKABLJ systems. At the reference densities, which are 1.5 for the KABLJ system and 1.0 for the RKABLJ system, each distribution was fitted with a smooth function (green dashed curves). For each system the scaling function $h(\rho) = A\rho^4 \pm B\rho^2$ was found by simulations of the equilibrium liquid at the reference density (see the main text for details). Based on these inputs and a dimensional analysis, the distributions at other densities are uniquely predicted (purple full curves).

steady state had been reached in which there is no memory of the initial state [41] and the statistics of the plastic flow events is strain independent. We then started probing the statistics of the stress drops $\Delta \sigma$, the strain intervals between mechanical instabilities $\Delta \gamma$, and the potential-energy drops ΔU . The algorithm used allows one to single out the drops to a high precision; thus, the instabilities are determined up to a resolution of $\sim 10^{-6}$ in strain. For each system studied statistics from about 20 000 plastic events were collected over 100 independent realizations in the steady state.

The probability distributions of potential-energy and stress drops are shown in Fig. 2 for different densities. The lowest density simulated for the KABLJ system is 1.2, corresponding to a slightly negative pressure. For the RKABLJ system we studied almost two decades of densities (pressure never becomes negative in a purely repulsive system).

In order to understand the scaling properties of the probability distributions we refer to the fact that both systems have strong virial potential-energy correlations [61,63]. As shown recently, this implies that they have “hidden scale invariance” [26,27] in the sense that the potential energy U as a function of the collective position vector $\mathbf{R} \equiv (\mathbf{r}_1, \dots, \mathbf{r}_N)$ obeys the following approximate identity:

$$U(\mathbf{R}) \cong h(\rho)\tilde{\Phi}(\tilde{\mathbf{R}}) + g(\rho). \quad (1)$$

Here, $\tilde{\mathbf{R}} \equiv \rho^{1/3}\mathbf{R}$ is the dimensionless “reduced” collective position vector, $h(\rho)$ and $g(\rho)$ are functions of density, both of dimension energy, and $\tilde{\Phi}(\tilde{\mathbf{R}})$ is a state-point-independent dimensionless function. Equation (1) defines a generalized, approximate scale invariance because the function $\tilde{\Phi}(\tilde{\mathbf{R}})$, which determines structure and dynamics of the equilibrium as well as nonequilibrium system [27], is scale invariant in the sense that it is unchanged for a uniform scaling of all particle coordinates.

According to Eq. (1) a change of density to a good approximation simply results in a linear, affine overall scaling of the high-dimensional potential-energy surface. For a system in thermal equilibrium this can be compensated by adjusting the temperature. This fact is the basis of the existence of the so-called isomorphs, which are curves in the thermodynamic phase diagram given by $h(\rho)/T = \text{Const.}$ along which structure and dynamics of equilibrium systems in properly reduced units are invariant to a good approximation [26,64].

Whereas the function $h(\rho)$ determines the energy scale for the dynamics at a given density, $g(\rho)$ just gives an additive constant to the potential energy and plays no role for structure and dynamics (this function contributes, of course, to the pressure and its density dependence and thus to the equation of state). Since $\tilde{\Phi}(\tilde{\mathbf{R}})$ is density independent, by dimensional analysis [65] Eq. (1) implies that for different densities the distribution

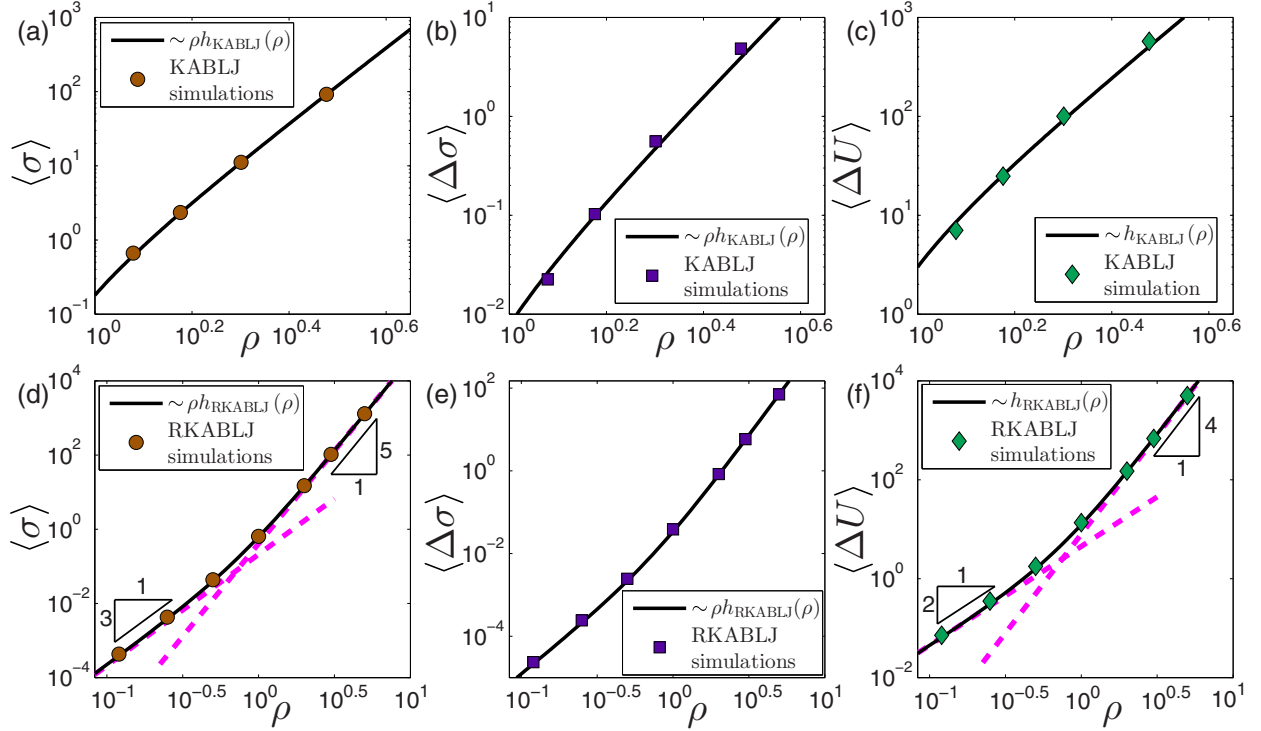


FIG. 3. (Color online) Mean steady-flow observables as functions of density for the KABLJ (top panels) and RKABLJ (bottom panels) systems. The continuous curves are proportional to $h(\rho)$ or $\rho h(\rho)$ for quantities having units of energy or stress, respectively. The purple dashed lines in two of the subfigures give examples of the predictions of asymptotic power-law scaling functions $h(\rho) \propto \rho^\gamma$. Previous effective scaling theories [44] predicted power laws corresponding to density-independent scaling exponents, which would imply the purple dashed lines to coincide. The deviations from simple power-law scaling are most pronounced for the RKABLJ system.

of flow-event potential-energy drops, $p(\Delta U)$, must be of the form $p(\Delta U) = F_U[\Delta U/h(\rho)]/h(\rho)$ in which the function F_U is density independent. Likewise, since stress has dimension energy per volume, the distribution of shear-stress drops must be of the form $p(\Delta\sigma) = F_\sigma\{\Delta\sigma/[\rho h(\rho)]\}/[\rho h(\rho)]$. In order to test these predictions we proceeded as follows.

First, the functions $h(\rho)$ for the KABLJ and RKABLJ systems were determined by making use of the fact that for a system with pair potentials of the form $v(r) = \sum_n \varepsilon_n (r/\sigma)^{-n}$, one has $h(\rho) = \sum_n \alpha_n \varepsilon_n (\rho \sigma^3)^{n/3}$ for suitable constants α_n [61,66]. The present case involves two inverse power-law terms, so the two $h(\rho)$ functions each have two parameters. Since in both cases the inverse-power-law exponents involved are $n = 12$ and $n = 6$, it follows that $h(\rho) = A\rho^4 \pm B\rho^2$. The overall scaling of $h(\rho)$ is irrelevant for the physics, however, so only a single parameter needs to be fixed for each system, e.g., B/A . This was done by simulations of the equilibrium liquid at a reference density by equating the expression for the density-scaling exponent, $\gamma = d \ln h/d \ln \rho$ [26,61,66], to its canonical-ensemble fluctuation expression $\gamma = \langle \Delta W \Delta U \rangle / \langle (\Delta U)^2 \rangle$ calculated from an NVT simulation [64]. As reference densities we chose $\rho = 1.5$ for the KABLJ system (resulting in $\gamma = 4.65$ and $B/A = 0.55$) and at $T = 1.0$ for the RKABLJ system (resulting in $\gamma = 3.29$ and also $B/A = 0.55$). Once $h(\rho)$ has been determined for each system, as mentioned above the plastic flow-event statistics at the reference density—marked by the

green dashed curves in Fig. 2—uniquely predict those at the other densities (purple full curves).

As a further investigation of the scaling properties implied by hidden scale invariance, Fig. 3 shows the average stress $\langle \sigma \rangle$, the average stress drop $\langle \Delta \sigma \rangle$, and the average potential-energy drop $\langle \Delta U \rangle$, as functions of density for the two systems. The full curves are the predictions from dimensional analysis: $\langle \sigma \rangle \propto \rho h(\rho)$, $\langle \Delta \sigma \rangle \propto \rho h(\rho)$, and $\langle U \rangle \propto h(\rho)$. For each quantity the proportionality constant was found from a reference-density simulation. The expression $\gamma = d \ln h/d \ln \rho$ for the approximate exponent defined by $h(\rho) \propto \rho^\gamma$ allows one to predict γ 's density dependence. The purple dashed lines in two of the subfigures of Fig. 3 give the low- and high-density power-law limits, respectively, which correspond to ordinary power-law scaling laws.

The scaling properties identified above allow one to compare different systems' flow-event statistics. Figure 4 shows the data for the KABLJ and RKABLJ systems, supplemented with data for the average shear displacement between flow events. The figure also includes data for a binary inverse-power-law (IPL) system with pair potentials $v(r) \propto r^{-10}$, for which the above-discussed scaling is exact and trivial with $h(\rho) \propto \rho^{10/3}$. These three quite different systems have very similar flow-event properties. We interpret this as reflecting a manifestation of the so-called quasiuniversality that was previously only discussed for equilibrium liquids (see, e.g., Refs. [57,67] and references therein). Thus, it has been known for some time that for simple liquids like inverse-power law systems, LJ systems, etc., the radial distribution functions are virtually

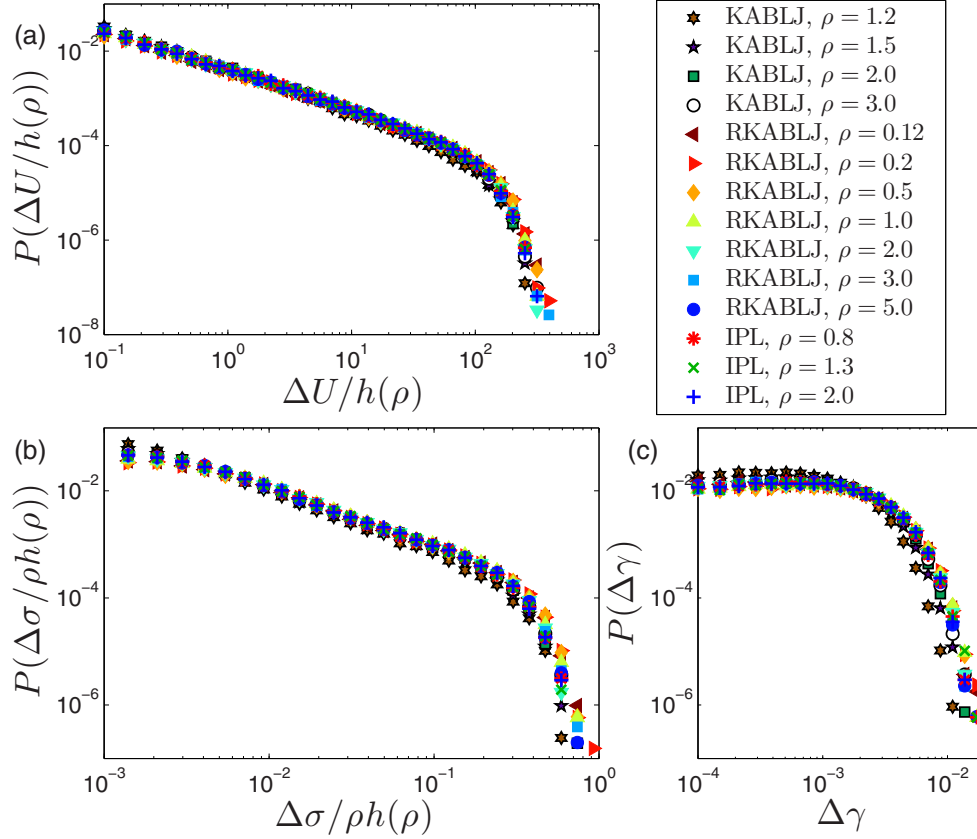


FIG. 4. (Color online) Distributions of dimensionless energy drops $\Delta U/h(\rho)$, stress drops $\Delta\sigma/[\rho h(\rho)]$, and strain intervals $\Delta\gamma$ for the KABLJ, RKABLJ, and $n = 10$ inverse power law (IPL) systems. This figure demonstrates quasiuniversality of the plastic flow-event properties. Note that this is not an empirical collapse; no adjustable parameters were involved.

indistinguishable [68,69]. More recently, quasiuniversality was extended to include simple liquids' dynamic properties [70,71], and very recently a theory based on the exponentially repulsive pair potential EXP has been devised explaining quasiuniversality for simple liquids [57].

In the hidden-scale-invariance language, quasiuniversality at thermal equilibrium implies that the function $\Phi(\mathbf{R})$ in Eq. (1) is quasiuniversal. In turn, this implies quasiuniversality of the flow-event statistics. Interestingly, in the related context of the jamming transition, Arevalo and Ciamarra very recently also reported quasiuniversality of flow-event properties close to jamming [72].

Which systems in the real world may have quasiuniversal behavior? Previous studies focused on liquid systems, for which it has been found that most or all van der Waals bonded systems and metals are in the “Roskilde-simple” class of systems with strong virial potential-energy correlations [27–36], i.e., obeying hidden scale invariance. In contrast, systems dominated by directional bonds like covalently bonded and hydrogen-bonded systems are generally not Roskilde-simple [27,73]. The microscopic basis is that systems with pair potentials that can be approximated by an inverse power-law term ($\propto r^{-n}$) plus a constant plus a linear term ($\propto r$) obey hidden scale invariance [27,73,74]. There is evidence that Roskilde-simple atomic systems, i.e., pair-potential systems with hidden scale invariance, also obey quasiuniversality [26,27,57]. Based on this we conjecture that most or all

metallic glasses exhibit quasiuniversality for their flow event properties. It is difficult to probe the statistics of individual flow events in experiments, of course, but for instance the ratio between the yield stress and the steady-state stress—a readily observable quantity—is also predicted to be quasiuniversal for glasses prepared according to the same thermal history.

The hard-sphere (HS) system has served well over the years as a generic simple model system, also for the glass phase via the (fairly well-defined) random close packing of spheres. However, the sort of scale invariance discussed above does not apply for the HS system, for which there is no analog of the scaling function $h(\rho)$. Rather, one may think of the HS system as the limit of inverse-power-law systems with exponents $n \rightarrow \infty$. Reference [57] argues that one can replace the HS system as the generic liquid system by the exponentially repulsive EXP pair-potential system, which has the advantage of being analytic and, moreover, gives a precise characterization of which (single-component) systems are quasiuniversal and which are not.

The implication of the above is that one may not only speak of Roskilde-simple liquids (systems with hidden scale invariance [73]), but also of Roskilde-simple glasses formed from such liquids. More work is needed to clarify the detailed consequences of this, however, since a glass is not uniquely determined by density and temperature but by the entire thermal history prior to glass formation.

In summary, we have shown that properties at a single density provide enough information to predict the plastic flow-event properties at arbitrary densities for the model systems studied. This consequence of hidden scale invariance was confirmed by simulations of the KABLJ and the RKABLJ systems. The resulting scaling of flow event properties reveal quasiuniversality of flow event properties, indicating that for

a large class of systems plastic flows may be simpler than previously thought.

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